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Crack opening profiles of indentation cracks in normal and anomalous glasses

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Abstract

A comparative Vickers indentation study is made of two glasses, soda-lime and borosilicate. Indentations in the two glasses reveal substantially shorter radial cracks in the borosilicate, even though toughness values measured by conventional (double-cantilever beam) methods are similar in the two glasses. Here, indentation toughness is measured in two ways: by optical measurement of the indentation crack lengths (ICL) and by atomic force microscopy (AFM) measurement of crack-opening displacements (COD) in the near-crack-tip regions. The ICL measurements indicate artificially high values for the borosilicate relative to the soda-lime, consistent with previously documented indentation results. The COD measurements indicate similar values for the two glasses, in line with expectations from the independent determinations. In the case of soda-lime glass, the COD and ICL values are mutually consistent. In the case of borosilicate, the COD and ICL values differ widely, indicating "anomalous" indentation behavior, typical of glasses with open, network-former structures. It is concluded that the COD route provides more reliable evaluations of intrinsic toughness, albeit at some expense in experimental simplicity. Residual elastic–plastic contact stresses responsible for driving the radial cracks, deconvoluted from COD measurements over the entire radial crack lengths, are shown to be significantly smaller in the borosilicate relative to soda-lime, indicative of a compaction rather than volume-conserving contact deformation mode.

Keywords: Indentation toughness; Crack-opening displacement; Anomalous glass; Residual stress

1. Introduction

Because of their relative simplicity, Vickers indentations are widely used to evaluate the toughness properties of brittle materials [1–6]. Toughness is most easily estimated from the length c of surface radial cracks around indents as a function of load P according to the relation

$$K_c = \chi P/c^{3/2}, \tag{1}$$

representative of center-loaded penny-like cracks, with coefficient

$$\chi = \xi (E/H)^{1/2}, \tag{2}$$

where E is the Young's modulus, H is the hardness and ξ is another coefficient [7,8]. The coefficient χ characterizes a residual elastic–plastic contact field, which drives the radial cracks during [7] and even after [9,10] unloading. It is the residual component of the contact stress field that determines the ultimate radial crack size in Vickers indentations. The indentation crack length (ICL) method of toughness determination [3] is now widely used by the brittle materials community for small specimens as a simple alternative to conventional small-crack fracture techniques like short-crack in flexure and single-edge v-notched beam [11,12].

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The quantity ξ is constant for a given indenter geometry provided the main assumption underlying the derivation of Eq. (2), that volume is conserved within the hardness plastic zone, is satisfied [8]. In this sense most brittle materials, including a wide range of network-modified glasses, behave in a "normal" manner – their deformation is shear activated and volume is indeed conserved. However, there are other brittle materials, especially network-forming glasses, that densify or compact below the indenter – such materials are said to behave in an "anomalous" manner [13]. Fused silica and borosilicate glasses fall into this latter category. Materials that densify are characterized by substantially diminished residual stress intensities at indentation sites, with consequently reduced radial crack lengths. Thus radial crack measurements in anomalous glasses can lead to gross overestimates of fracture toughness [13]. The quantity ξ in Eq. (2) is then no longer material-independent, placing the universality of Eq. (1) in question.

Another, more complex way of evaluating toughness from indentations is to measure crack-opening displacements (COD) close to the crack tips in high magnification, e.g., by scanning electron microscopy (SEM) [14–17] or atomic force microscopy (AFM) [18]. Toughness is determined from the crack profile. COD methods require only that the crack remains in quasi-equilibrium, i.e., on the verge of extension, which is the case immediately after indentation with sharp indenters [7]. Such COD methods provide a means for examining intrinsic toughness properties in a direct manner.

In this paper we compare ICL and COD toughness evaluations for two archetypical normal and anomalous glasses, soda-lime and borosilicate, for Vickers indentations. We demonstrate the greater reliability of the COD technique as a means of intrinsic toughness determination, albeit at considerable expense in simplicity. The COD measurements can be extended over the entire radial crack lengths so that the nature and intensities of the underlying crack driving forces – in this case the residual contact stresses – can be deconvoluted. Such deconvolutions confirm a much reduced residual stress intensity in the anomalous glass. Thus, taken together, ICL and COD measurements, in addition to providing toughness values, enable basic evaluations of residual contact stress fields.

2. Experimental

Commercially available soda-lime glass (Euroglas Haldensleben, Dammmühlenweg 60, Haldensleben, Germany) and borosilicate (Duran, Schott-Rohrglas GmbH, Potfach 101152, Bayreuth, Germany) glass were selected as representative normal and anomalous silicate glasses, respectively [13,19]. The chemical composition of the two glasses is given in Table 1. The chief difference between the two compositions is the large component of network modifiers (Na, Ca) in the soda-lime and network formers (B) in the borosilicate, the latter typified by a relatively "open" structure [13].

Table 1
Chemical composition of the glasses under investigation (wt%)

Glass	SiO_2	B_2O_3	Al_2O_3	CaO_2	MgO	Na_2O
Soda-lime	72	–	0.3	9	4	14
Borosilicate	81	13	2	–	–	4

Indentations were performed with a Vickers diamond pyramid at a dwell time of 15 s at ambient temperature (25 °C) and relative humidity (47%). Loads of 9.8, 29.4, 49.0, 78.5, and 98.1 N were applied in the case of soda-lime glass, and 9.8, 29.4, and 49.0 N in the case of borosilicate glass (higher loads in the borosilicate led to chipping). The indentations were allowed to sit in laboratory atmosphere for two days to allow the radial cracks to extend subcritically under the influence of moisture to a "steady state" condition before measuring the crack dimensions [3,9,10]. Typical surface views at common load $P = 49$ N in Fig. 1 reveal the classical crack pattern, but with considerably smaller radial arm lengths and exaggerated shear-fault structure in the borosilicate glass [13].

An idealized schematic of the indentation geometry is shown in Fig. 2. Radial crack lengths c and hardness impression half-diagonals a were measured by optical microscopy in Nomarski contrast. COD displacements u were measured at positions r along the crack lengths using an atomic force microscope (AFM, Digital Instruments, Veeco Metrology Group, Santa Barbara, CA) in tapping mode, with Si cantilever tips (accuracy $\pm$ 15 nm). Surface areas 10×10 μm^2 were scanned at 0.5 Hz in this mode. Displacements at positions $x = c - r$ close to the crack front were measured to higher accuracy ($\pm$5 nm) over surface areas 3×3 μm^2 using super cone cantilevers with much sharper (5–10 nm radius) tips.

Elastic moduli E were measured for each glass by a routine impulse excitation method (IMCEnv, B-3590 Diepenbeek, Belgium), and hardness H directly as $H = P/2a^2$.

3. Toughness results

Indentation crack length toughness evaluations were made from Eq. (1) in conjunction with a fixed coefficient $\xi = 0.016$ in Eq. (2), calibrated from tests on normal materials [3]. These determinations are shown in Table 2, along with K_c values from independent double-cantilever-beam (DCB) tests in vacuum [19]. Two features are evident: first, the indentation values for soda-lime glass are lower than from the vacuum DCB tests, consistent

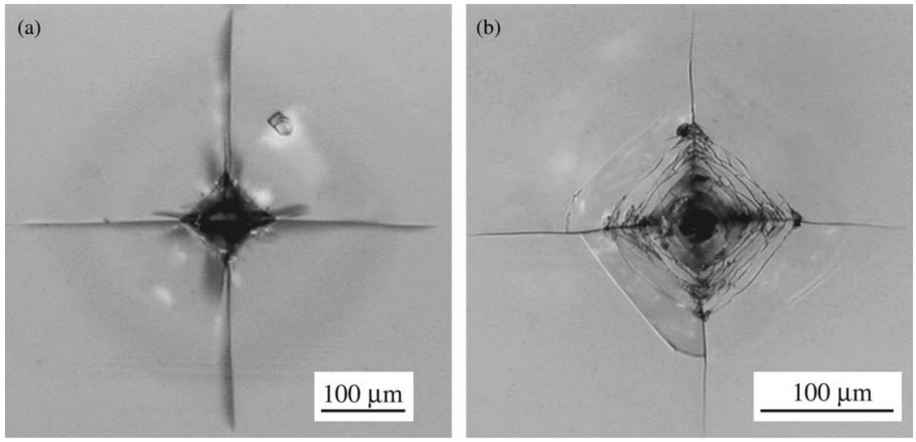

Fig. 1. Optical micrographs of Vickers indents made at load $P = 49$ N in (a) soda-lime and (b) borosilicate glass. Note different magnifications in two images.

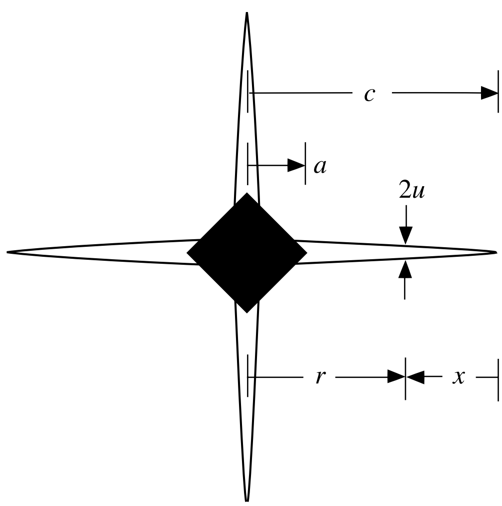

Fig. 2. Schematic representation of the radial crack system around a Vickers indent.

Table 2
Toughness of soda-lime and borosilicate glass determined from ICL and COD measurements, ambient atmosphere (measurement uncertainty bounds ± 0.02 MPa $m^{1/2}$)

Toughness (MPa $m^{1/2}$)	Soda-lime	Borosilicate
DCB (vacuum) [19]	0.75	0.76
Indentation crack length	0.55	1.2
Indentation COD	0.47	0.49

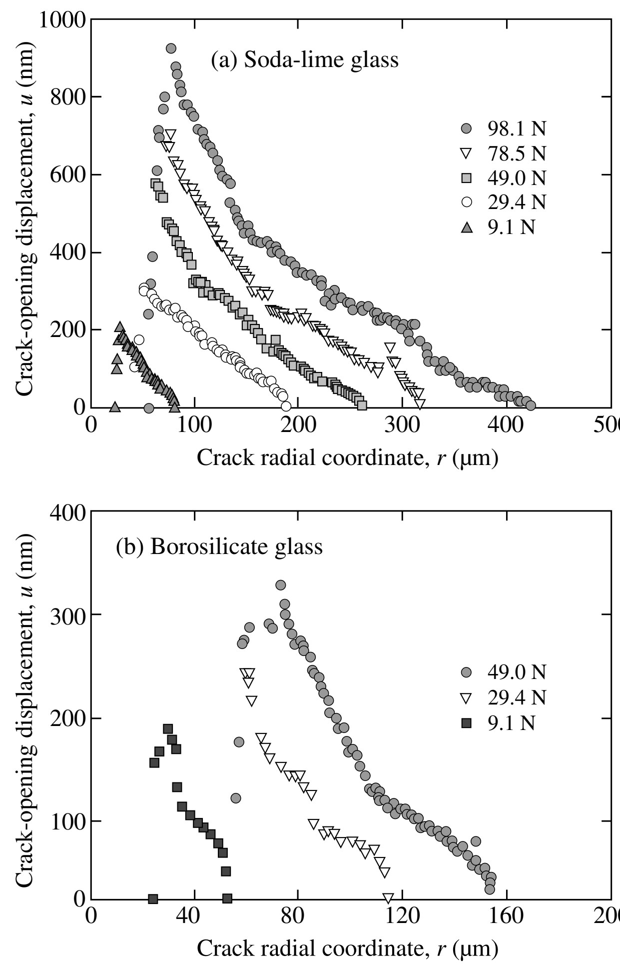

Fig. 3. Crack-opening displacements $u(r)$ for Vickers indentations in (a) soda-lime and (b) borosilicate glass, at loads indicated. Note different scales for the two materials.

with some reduction in effective toughness with post-indentation crack extension in the moist air; and second, the ICL toughness is considerably higher for the borosilicate relative to the soda-lime glass, consistent with a reduced coefficient ξ for anomalous glass.

Measured COD data $u(r)$ are plotted in Fig. 3 for soda-lime and borosilicate glasses, at the indentation loads P indicated. The general shape of the profile is the same in all cases for each material. Note, however, that the COD values are smaller in the borosilicate than in the soda-lime at any given load. Note also that the cracks extend partially into the compressive hardness

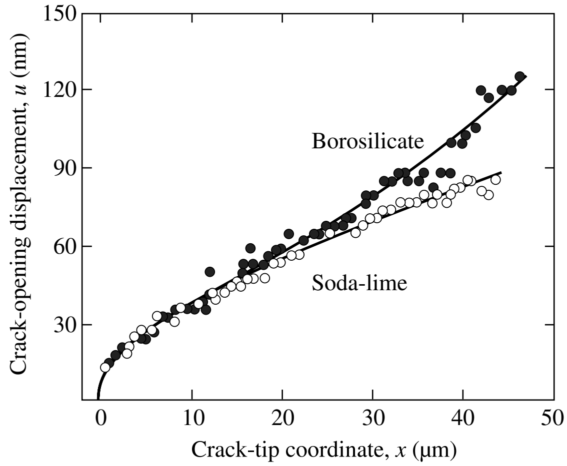

Fig. 4. Crack-opening displacements $u(x)$ in the tip near-tip field for Vickers indentations in soda-lime glass (unfilled symbols) and borosilicate glass (filled symbols), at common load $P = 49$ N. Curves are data fits to Eq. (3).

zone at $r < a$ (corresponding to the position of maximum COD). At $r > a$ the $u(r)$ curves are concave upward over the discernible data range in Fig. 3, consistent with a centrally loaded penny crack. A more detailed analysis of these profiles across the full indentation coordinate r will be given elsewhere [20].

More accurate near-tip COD measurements plotted as functions of crack-tip coordinate x in Fig. 4 for a common load $P = 49$ N indicate a reversal of this curvature in the regions immediately behind the crack tip. This is consistent with a near-tip displacement field of form [6]

$$u(r) = (8x/\pi)^{1/2}(K_c/E')x^{1/2} + k_1x^{3/2} + k_2x^{5/2} + \cdots, \quad (3)$$

where $E' = E/(1-2v)$, v is the Poisson's ratio, and k_1 and k_2 are higher-order stress-intensity factor terms. The second and third terms on the right of Eq. (3) are included to allow for deviations from ideal parabolic contours at large x, especially pronounced for the relatively small cracks in the borosilicate glass. Values of toughness K_c from fits of Eq. (3) to the data in Fig. 4 (solid curves) are included in Table 2. Note that these values are the same for the two glasses within experimental scatter, as may be expected from the independent DCB values. However, whereas the COD K_c values are comparable to those from ICL determinations for soda-lime glass, they differ significantly for borosilicate glass.

4. Residual contact stress analysis

The discrepancy between toughness values evaluated from crack length and COD measurements for the anomalous borosilicate glass warrants a closer look in terms of residual stress intensities associated with the plastic component of the indentation contact. At distances r the COD function has the integral form [14,16]

$$u(r) = (1/E')\int_r^c h(c',r)\left\{\int_0^{c'} \sigma(r')h(c',r')\,\mathrm{d}r'\right\}\mathrm{d}c', \quad (4)$$

where $\sigma(r')$ is the pre-crack residual stress acting at a location r' and h is a weight function. For the purposes of analysis, we use the weight function for open penny cracks with a radial stress distribution [6]

$$h(c,r) = (2r/\pi^{1/2})/[c(c^2-r^2)]^{1/2}. \quad (5)$$

With this function, the stresses $\sigma(r)$ may then be deconvoluted numerically from Eq. (4).

Plots of $\sigma(r)$ are shown in Fig. 5 for the two glasses as a function of relative crack coordinate r/a, for several indentation loads. The solid lines are best fits to the power-law function [21,22]

$$\sigma(r) = \sigma_R(a/r)^3, \quad (6)$$

where σ_R is the value of σ at $r = a$. This function fits the data within the scatter over the range r for soda-lime glass, but shows some deviations for borosilicate glass. Notwithstanding such deviations in the latter instance, the relative values of residual stress intensity for the two glasses, $\sigma_R = 1.24$ GPa for soda-lime and $\sigma_R = 0.55$ GPa for borosilicate, show significant differences. The relative values of σ_R are consistent with about a factor of 3 difference between normal and anomalous glasses from earlier stress birefringence measurements [13]; the high absolute values (~GPa) emphasize the intensity of the residual contact deformation.

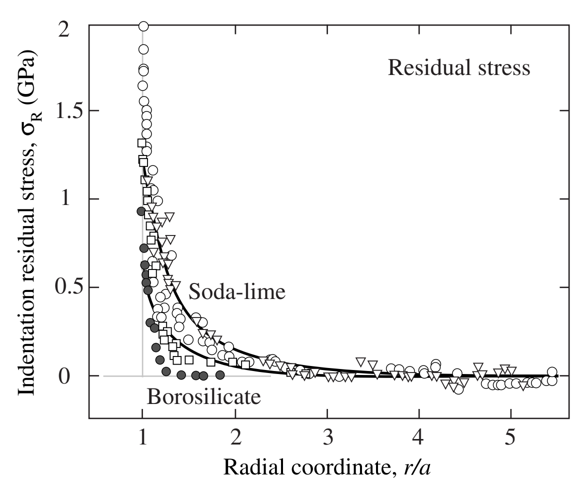

Fig. 5. Residual stresses in soda-lime glass (unfilled symbols) and borosilicate glass (filled symbols), data for several indentation loads, from deconvolution of data in Fig. 3.

5. Discussion

Indentation measurements on soda-lime and borosilicate glasses confirm the findings of an earlier study [13] that the latter glass behaves in an "anomalous" manner. Whereas independent, long-crack (DCB) toughness tests indicate the two glasses to have essentially similar K_c values, determinations from Vickers radial crack length measurements using a coefficient $\xi = 0.016$ in Eqs. (1) and (2) indicate a greatly exaggerated value for the borosilicate (Table 2). Recall that the evaluation $\xi = 0.016$ was originally determined by calibration against "normal" materials with independently known toughnesses [3]. In order for the borosilicate glass to render a toughness similar to soda-lime, a much smaller coefficient $\xi = 0.006$ would have to be used. This discrepancy is attributable to the different levels of residual elastic–plastic contact stresses that drive the radial cracks. In soda-lime glass, the plastic component of contact deformation is shear-driven, with conservation of material volume; in borosilicate, the plastic component is compression-driven, with resultant material densification. The latter deformation mode is less effective in expanding the surrounding elastic material outward upon removal of the indenter [8]. The radial crack lengths are consequently smaller in the anomalous material, with the resultant overestimate in toughness from ICL measurements.

Crack-opening displacement measurements in the vicinity of the crack tip (Fig. 4) provide an alternative estimate of toughness. These measurements make use of the near-parabolic contour of the crack walls in the near-tip region [6,14,15]. The toughness evaluations are similar for the two glasses (Table 2), within the scatter in data, consistent with the DCB trend. Hence the COD method provides more reliable estimates of the intrinsic material toughness. On the other hand, the measurements are more onerous, generally requiring recourse to high magnification, high accuracy observation techniques like SEM or AFM. Generally, the two measurement methods, crack length and COD, need to be taken together in order to distinguish between normal and anomalous behavior in any glass system. In polycrystalline ceramics with *R*-curves, COD evaluations can be used to determine both the intrinsic crack-tip toughness and, ultimately, the extrinsic shielding toughness (from crack-wall bridging [16] and process-zone toughening [18]). Again, intercomparisons between different measurement techniques are called for.

Crack-opening displacement measurements over the entire radial crack lengths enable quantitative evaluations of residual stresses at the contact site. The present results indicate significantly higher stress intensities in the normal relative to anomalous glass (Fig. 5), $\sigma_R =$ 1.24 GPa for soda-lime and 0.55 GPa for borosilicate. This ratio is consistent with previous evaluations using polarized light [13].

The methodology presented in this study should prove useful as a means of characterizing the deformation response of glasses and other brittle materials.

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References

[1] Lawn BR, Wilshaw TR. J Mater Sci 1975;10:1049.
[2] Evans AG, Charles EA. J Am Ceram Soc 1976;59:371.
[3] Anstis GR, Chantikul P, Marshall DB, Lawn BR. J Am Ceram Soc 1981;64:533.
[4] Chantikul P, Anstis GR, Lawn BR, Marshall DB. J Am Ceram Soc 1981;64:539.
[5] Cook RF, Pharr GM. J Am Ceram Soc 1990;73:787.
[6] Lawn BR. Fracture of brittle solids. 2nd ed. Cambridge: Cambridge University Press; 1993.
[7] Marshall DB, Lawn BR. J Mater Sci 1979;14:2001.
[8] Lawn BR, Evans AG, Marshall DB. J Am Ceram Soc 1980;63:574.
[9] Gupta PK, Jubb NJ. J Am Ceram Soc 1981;64:C112–4.
[10] Lawn BR, Jakus K, Gonzalez AC. J Am Ceram Soc 1985;68: 25–34.
[11] ASTM C 1421-99, Annual book of standards, vol. 15.01. West Conshocken, PA: American Society for Testing and Materials; 2001.
[12] Moon R, Bowman K, Trumble K, Rödel J. J Am Ceram Soc 2000;83:445.
[13] Arora A, Marshall DB, Lawn BR, Swain MV. J Non-Cryst Solids 1979;31:415.
[14] Rödel J, Kelly J, Lawn BR. J Am Ceram Soc 1990;73:3313.
[15] Seidel J, Rödel J. J Am Ceram Soc 1997;80:433.
[16] Fett T, Munz D, Seidel J, Stech M, Rödel J. J Am Ceram Soc 1996;79:1189.
[17] Pezzotti G, Muraki N, Maeda N, Satou K, Nishida T. J Am Ceram Soc 1999;82:1249.
[18] Meschke F, Raddatz O, Kolleck A, Schneider GA. J Am Ceram Soc 2000;83:353.
[19] Wiederhorn SM, Johnson H, Diness AM, Heuer AH. J Am Ceram Soc 1974;57:336.
[20] Fett T, Kounga AB, Rödel J. J Mater Sci Lett [in press].
[21] Hill R. The mathematical theory of plasticity. London: Oxford University Press; 1950.
[22] Johnson KL. Contact mechanics. London: Cambridge University Press; 1985.